'top of the figure', with the high dose rate and stops at the 'bottom' with a very low dose rate and low temperature. Along each trajectory the time is indicated with markers starting with 10^{-1} [a]. The 'distance' between the markers is a factor 10.

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THE THEORY OF RADIATION DAMAGE IN SALT CRYSTALS AND ROCKS, THE LEADING QUESTIONS AND UNDERLYING RESEARCH THESES

A. García Celma, L. Vons and H. Donker

ABSTRACT

Taking into account repository concepts for final disposal of radioactive waste in salt formations consisting of long vertical boreholes where the vitrified HLW–canisters devoid of an extra overpack are emplaced, a methodology was developed to prove or disprove some essential theses. Some of these theses constituted the basis of pessimistic estimations regarding the safety of these concept repositories. The theses refer to the accumulation of stored energy in the form of lattice defects in the crystals of the rock salt due to γ–irradiation. Following these estimations accumulation of crystal defects could bring about spontaneous explosive back reactions which would threaten the containment of the waste. This article shortly introduces these theses, explains the methodology applied to be able to prove or disprove them, summarily presents the experimental and theoretical results obtained following the described methodology and, after discussing the veracity of each of the theses concludes that no scientific basis has been found to assume that spontaneous explosive back reactions would take place in eventual repositories built according to the considered concepts.

1. INTRODUCTION

In 1989 an "International Test Plan" [Mönig et al., 1990] was formulated, which coordinated the different ways in which some remaining questions regarding radioactive waste disposal in rock salts would be tackled and which settled the responsibilities of the different partners. The "International Test Plan" document establishes a series of irradiation experiments directed to answer remaining questions regarding on the one hand the problem of gas development in repositories and on the other hand the problem of radiation damage development.

These experiments were planned to be partly carried out in various laboratories but mainly in the HAW (High–Active Waste) test field in the Asse salt mine, Remlingen, Germany. Information on the HAW–test field can be found in [Rothfuchs et al., 1988; Rothfuchs, 1995].

Part of the task of the ECN (Energieonderzoek Centrum Nederland, The Netherlands Energy Research Foundation), our task, was to study the radiation damage development and anneal in rock salt in collaboration with the Laboratori d'Investigació en Formacions Salines of the Barcelona Universtiy under contract by ENRESA (Empresa Nacional de Residuos Radioactivos de España Sociedad Anónima). The ECN, moreover enjoyed a fruitful collaboration with the U.U. (Utrecht University) where four staf members of the project have worked during all these years.

In the HFR (High Flux Reactor) at Petten two Gamma Irradiation Facilities containing rock salt samples were in operation, GIF A and GIF B. The gamma–irradiations performed with these facilities took place at a constant temperature (100°C). GIF A irradiations took place at variable and GIF B at approximately constant dose rates. Initially these facilities were build to monitor the irradiation experiments that were planned in the HAW test field. Due to repeated delays in (and definitive cancellation of) the emplacement of the radioactive sources in the HAW field, these complementary laboratory experiments were steadily extended.

Here we will give a summary of the leading questions in our work and of the way in which answers to them were sought, and eventually found, since 1989 up to 1994. The questions which remained to be solved at the start of this research will be first explained, then we will resume how we intended to find the answers to them in our methodology chapter and, after a summary of the results and some discussion, the most important conclusions will be given.

2. PROBLEM DEFINITION: THE BASIC THESES

The general question leading our research is : Will radiation damage in rock salt endanger the containment of the waste in radioactive waste repositories?.

To answer this question is not easy, because experiments for the length of time during

which gamma radiation above the natural background will be present in a repository are impossible to carry out. Experimentators have to choose between reproducing the total dose to be expected in a repository by using irrealistically high dose rates or to reproduce the dose rate and never reach the total dose.

Moreover, since dose rate and total dose do not relate linearly with the obtained radiation damage we have to rely on rather complex computer simulations based on theoretical models to predict the behaviour of a repository. To develop the computer simulations, the processes implied in radiation damage development have to be well understood. One of the best known theoretical models is that of Jain and Lidiard [Jain and Lidiard,1977]. Modifications to the Jain–Lidiard model had been introduced before we started this research [Lidiard, 1979; Van Opbroek and den Hartog, 1985], and some other modifications were suggested and/or implemented during these years. Most modifications and theory development during our research are either the work of the Groningen University [Groote and Weerkamp, 1990; Seinen et al., 1992; and Seinen, 1994] or are the result of our work [Soppe, 1993; Soppe and Kotomin 1994; Soppe and Prij, 1994 a, and 1994b; Soppe et al., 1994 and Donker et al., in prep]

At the beginning of our research little was known about the factors and/or functions that relate the experimentally obtained results, at high dose rates, with those expected under repository conditions, at low dose rates. Our research was therefore aimed at controlling the validity, for the case of a repository, of the thesis underlying the safety problems which could be envisaged as a result of radiation damage in rock salt. An essential part of the experimental programme consisted of narrowing the gap between the experimental irradiations and the prospective repository irradiations of rock salt in what respects dose rates and total doses. Figure 1 shows that the conditions of irradiation in the HFR experiments are nearer to repository conditions than ever was the case in a long lasting laboratory experiment. The theses considered can be resumed as follows:

a) all other conditions being constant, low dose rates of gamma rays produce more damage than high dose rates until a maximum is reached and this efficiency decreases again. Repository conditions are thought to lie in the dose rate regime where the efficiency increases. This thesis stems from theoretical models [Van Opbroek and Den Hartog, 1985; Groote and Weerkamp, 1990].

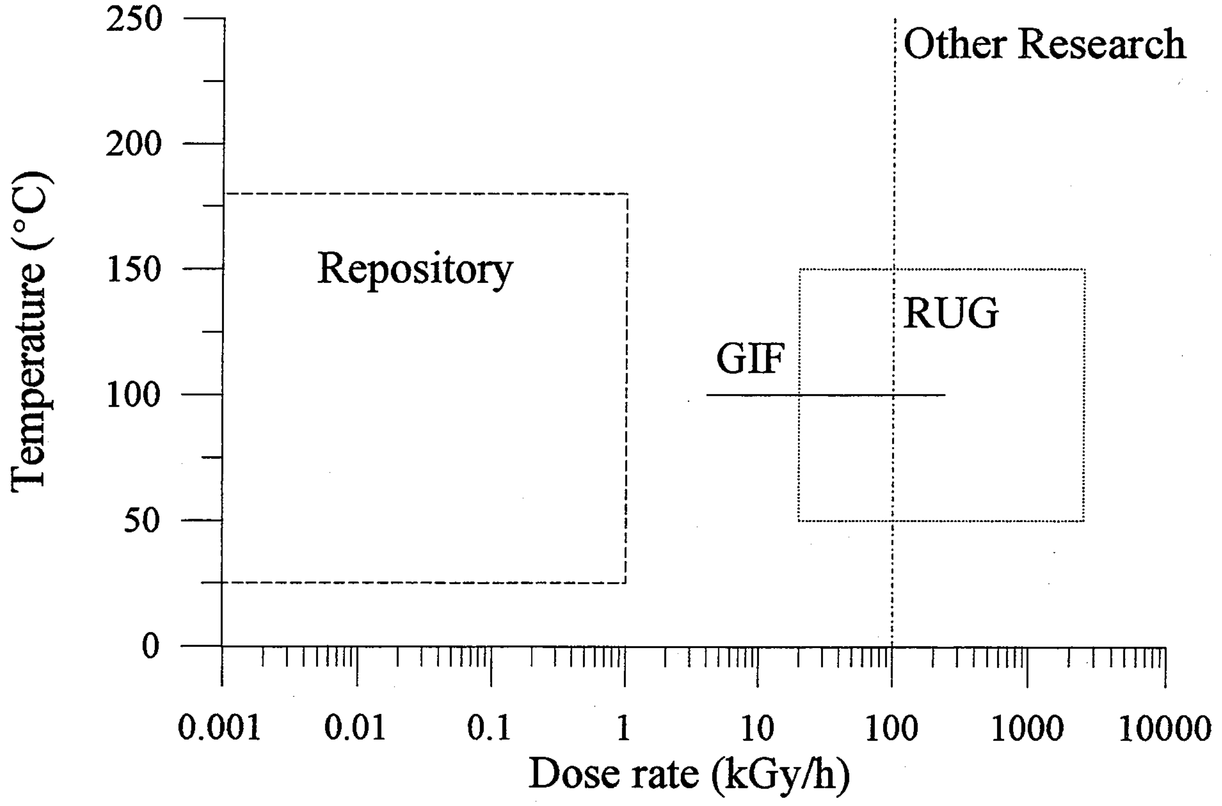

Figure 1: *Temperature and dose rate range of the experiments performed at the Groningen University (RUG), the GIF A&B experiments at the ECN and those to be expected in a repository.*

b) crystal defects –either mechanical (dislocations), or chemical (impurities)– enhance the efficiency of radiation damage development. All other conditions constant, impure and /or strained crystals ought to develop more damage than pure undeformed crystals. According to this, and to the fact that natural rock salts are always deformed and impure, natural rock salts ought to contain more stored energy than the pure single crystals used in most experiments. This thesis derives from interpretations of experimental work performed by [Groote and Weerkamp, 1990; Compton ,1957; Ikeda and Yoshida, 1967; Lèvy et al.,1980, and 1981; Den Hartog ,1988 and Den Hartog et al., 1990].

c) radiation damage in a repository will not reach a saturation level but will grow until a percolation limit is reached and an instantaneous back reaction takes place. This thesis was poned at the Groningen University [Groote and Weerkamp, 1990; Seinen et al., 1992; Den Hartog et al., 1990 and Weerkamp et al., 1994] and is based on the interpretation of experimental results on NaCl doped with K and on the predictions of the original Jain–Lidiard model.

d) if the huge amounts of stored energy that were found in crystals irradiated at the Groningen University are taken into account "explosive back reactions" could threaten the integrity of a repository [Den Hartog et al., 1990; Prij, 1991 and number 2 and 23 in this volume]. This is based on rock mechanics calculations performed at the Solid State Physics Department of the Groningen University.

e) neither fluid assisted recrystallization, nor other sorts of creep mechanisms will act in any significant way during the active life of a repository. Therefore the influence of these mechanisms in reducing stress (and consequently, stored energy), although recognized when discussing the rheological properties of a repository [Spiers et al., 1986 and Urai et al., 1986] is absolutely ignored when discussing radiation damage.

Theory development from which these basic theses stem, had been mostly based on experiments on pure (or containing controlled amounts of impurities) undeformed single crystals irradiated at atmospheric pressure (or in vacuum). These crystals and conditions are, however, very different from the impure, polycrystalline and deformed rock salt irradiated at the lithostatic pressure expected to occur in a repository. Although we aimed at reproducing repository conditions as nearly as possible, samples such as pure undeformed monocrystals and irradiation of samples at atmospheric pressure, were nonetheless included in the experimental plan to form a link with existing theories and to allow us to countercheck the theses. To this end a variety of samples was irradiated in two sorts of experiments: GIF A experiments, which were planned to prove or disprove theses *c*, *d*, and *e*, and GIF B experiments which were planned to prove or disprove theses *a*, *b* and *e*.

3. METHODOLOGY

3.1. Irradiation experiments

All irradiation experiments (both GIF A and GIF B) were performed at the same and constant temperature of 100°C. The temperature of irradiation is a very important parameter in radiation damage development, the effect of temperature variation on radiation damage was however out of the scope of our study due to various reasons. The most important reason for excluding temperature variation of our research was that the limited amount of experiments which could be carried out forced us to fix a constant value for at least one of the important parameters. Another important reason was that it was planned to study the effect of temperature in radiation damage in the HAW–field experiment with samples identical to those irradiated by us and up to the same total dose, and last but not least the influence of temperature in radiation damage development had already been extensively studied in laboratory experiments [Lèvy et al.,1981 and Den Hartog, 1988]. Since in this last named studies it was concluded that the temperature at which irradiation was most efficient in producing radiation damage was that of 100°C, this temperature was chosen for all our laboratory experiments as a way to maximize the obtained damage.

In the GIF A experiments 40 samples were irradiated simultaneously at a variable dose rate (240 to 20 kGy/h in monthly cycles). Each month one of these samples was retrieved and analyzed. In a first experiment (GIF A0) a total dose of 567 MGy was reached, after which the equipment and salt samples contained in it were accidentally flooded with water from the cooling pool of the reactor due to a leak in a minitube. As a consequence the experiment had to be restarted. The restarted experiment, or second experiment (GIF A1) was successfully carried out for a period of 3 years and a total dose of 1200 MGy was reached. The total dose interval between the subsequently retrieved samples was approximately 30 MGy. The irradiated samples were Asse Speisesalz of the 800 m depth level, without added brine and at about atmospheric pressure.

In each GIF B experiment 16 samples of different composition were simultaneously irradiated up to one of the total doses expected in the HAW field experiment (0.02 to 44 MGy), but at a higher dose rate (4 – 15 kGy/h in GIF B versus the planned 0.01, 0.1 and 1 kGy/h for the HAW field) and at a constant temperature (100 °C in GIF B versus 85 to 150 °C in the HAW field).

Each sample, of those irradiated together in the GIF B experiments, differs from the others in one or more of the following factors: microstructure, composition, amount of added brine, or pressure. The samples were placed in separate holders which allowed pressures to be applied by means of gas bellows. The instrumentation was designed, tested and produced especially for this research [García Celma , 1991 and García Celma et al., 1991a].

The starting materials consisted of pure NaCl Harshaw (H) monocrystals, pressed pure NaCl powder (PP), synthetic rock salt (SS), and different kinds of natural rock salt : Speisesalz of the 800m level, Borehole polyhalitic salt, Borehole anhydritic salt and Polyhalitic salt (all provinient from the Asse Salt Mine, Remlingen, Germany) and Potasas del Llobregat salt (from the Potasas del Llobregat Mine, Sallent, Spain).

In GIF B three sets of experiments were performed. In the first set (GIF B1) the average dose rate was 15 kGy/h, in the second set (GIF B2) 4 kGy/h and in the third set (GIF B3) 15 kGy/h. The GIF B1 and B2 experiments were performed to be able to specifically answer the question on the enhanced efficiency of damage formation for low dose rates. The GIF B3 experiments were performed in order to check some controversial results of the GIF B1 experiments.

In the GIF B1 experiments (at 15 kGy/h) none of the samples was annealed prior to irradiation. In the GIF B2 experiments (at 4 kGy/h) the Harshaw monocrystals, which had been shown to contain variable amounts of stored energy, were annealed prior to irradiation [García Celma and Donker, 1994a]. In the GIF B3 experiments, annealed and non–annealed Harshaw crystals were irradiated simultaneously.

3.2. Performed analysis

The different aspects of the damage developed during the irradiation experiments described above were analyzed by the ECN–UU and the BU [De las Cuevas and Teixidor, 1992 and Pueyo et al., 1992] each on one piece of the same sample. The results were compared with the yields of the Jain–Lidiard model for each experiment. The irradiations, stored energy analyses, most of the microstructural analyses, the Jain–Lidiard calculations and further model development were carried out by the ECN–UU. Light Absorption analyses, measurements of hydrogen yields by solution of the samples in H_2O, thermogravimetry , geochemical characterization and microstructural analysis of the Potasas del Llobregat samples were performed at the Barcelona University.

Stored energy measurements were performed on the darkest material which could be selected from each sample after irradiation (recrystallization taking place during irradiation produces white crystal areas which do not contain stored energy [García Celma et al., 1988 and 1993a]. Small sized second phase minerals and perhaps little pieces of recrystallized material may still have been present in the samples prepared for measurements. The results of stored energy determinations in the darkest pieces of irradiated samples were corrected for the original stored energy contents of non–irradiated samples (which was measured in equivalent non–irradiated samples) and for thermal effects due to impurities, when indicated [García Celma and Donker, 1994a]. These corrected results were used in interpretation, although polycrystalline rocks evidently have less stored energy since they also include recrystallized parts and impurities. The most common impurity in our samples is polyhalite which, when heated, loses its compositional water in an endothermal process (absorbing heat and lowering the temperature) [Jockwer, 1981 and García Celma, 1991b].

4. EXPERIMENTAL RESULTS

4. 1. GIF A results

Figure 2 gives the evolution of stored energy with increasing total dose found for the GIF A experiments. A saturation level of radiation damage was sought for, and has been found at about 140 J/g (corresponding to approximately 1.7 mol% of the salt being decomposed). In Fig. 2 the measured stored energy values are also compared with the results of model calculations using the modified Jain–Lidiard model [Van Opbroek and Den Hartog, 1985] and the extended Jain–Lidiard model by Soppe [Soppe, 1993]. In both models some parameters were modified as based on our latest insights [Donker et al, in prep]. In Fig. 2 it can be observed that both models predict more damage than experimentally observed. The conversion factor used for comparing stored energy and amount of defects is 5 eV/defect corresponding to 82.5 J/g per mol% damage as discussed in [Donker et al, in prep].

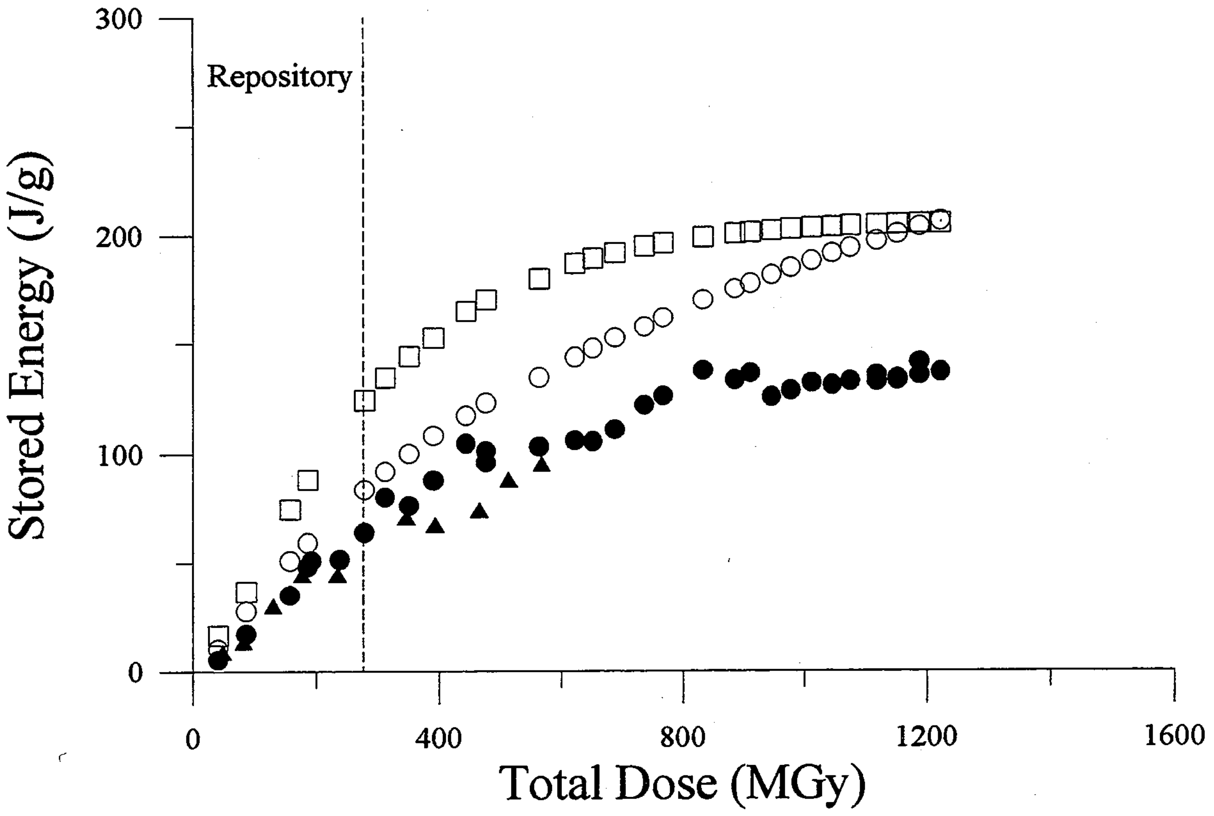

Figure 2: *Comparison of measured stored energy values on Sp–800 samples irradiated in GIF A0 (triangles) and GIF A1 (full circles) with model calculations using the modified Jain–Lidiard model (squares) and with model calculations using the extended Jain–Lidiard model by Soppe (open circles). The model calculations have only been performde for the GIF A1 experiments. The dashed line indicates the maximum total dose expected for a repository (see text).*

4. 2. GIF B results

The results of the stored energy measurements performed on the Harshaw crystals and Sp–800 samples irradiated in the GIF B experiments are shown in Fig. 3 to 5.

In the GIF B1 experiments we have observed that the samples, which were irradiated at a low and constant dose rate (15 ± 5 kGy/h), show, after an initial increase of the stored energy and a maximum value at about 2.6 MGy, a decrease of stored energy with increasing total dose. At a total dose between 5 and 15 MGy a minimum is reached after which the stored energy increases again with increasing total dose (see Fig. 3 and 4). Below total doses of 15 MGy a comparison of our stored energy results with the results of the Light Absorption measurements shows that the irradiated samples yield higher stored energy levels than could be attributed to colour centres [García Celma, 1993 ; García Celma and Donker, 1994b and García Celma et al., 1992 and 1993b]. The initial increase and the ensuing decrease of stored energy was therefore ascribed to the development of dislocations and their anneal, also because the stored energy peaks were observed at a higher temperature than the colloid anneal peak in the thermograms of the Harshaw crystals irradiated to higher doses [García Celma and Donker, 1994a]. Moreover, the development of low energy dislocation arrangements has been shown by means of microstructural analysis [García Celma and Donker, 1994b]. However, the results of the GIF B3 experiments shed some doubts on this interpretation.

In the GIF B3 experiments (also performed at a dose rate of 15 kGy/h) the stored energy observed for samples irradiated up to total doses of 11 MGy is at an approximately constant value of about 2 J/g (see Fig. 5). Not only is this value much lower than those observed in the GIF B1 experiments, also the initial rise and ensuing decrease of stored energy as found in the GIF B1 experiments is not observed. The experimental conditions under which both sets of experiments were performed were similar, therefore the reason for the observed differences is not yet very clear. At this moment we think that they might be caused by surface effects. Jiménez de Castro and Álvarez Rivas [Jiménez de Castro and Álvarez Rivas,1990] have reported that samples cut from the surface of their irradiated crystals show an enhanced stored energy in the temperature region above 300 °C, compared to the samples cut from the interior of the same irradiated crystal. In the GIF B3 experiment we are certain that all the samples used for stored energy analysis were cut from the interior of the irradiated cylinders or tablets while this was most probably not the case in the GIF B1 experiments.

Another cause for the observed differences between GIF B1 and GIF B3 results might be the containment of the samples. In the GIF B1 experiments the samples were wrapped in silver foil, while in the GIF B2 and B3 experiments they were contained in gold. We have shown that the silver foil had partly reacted with chlorine from the salt and that the folds of the silver foil

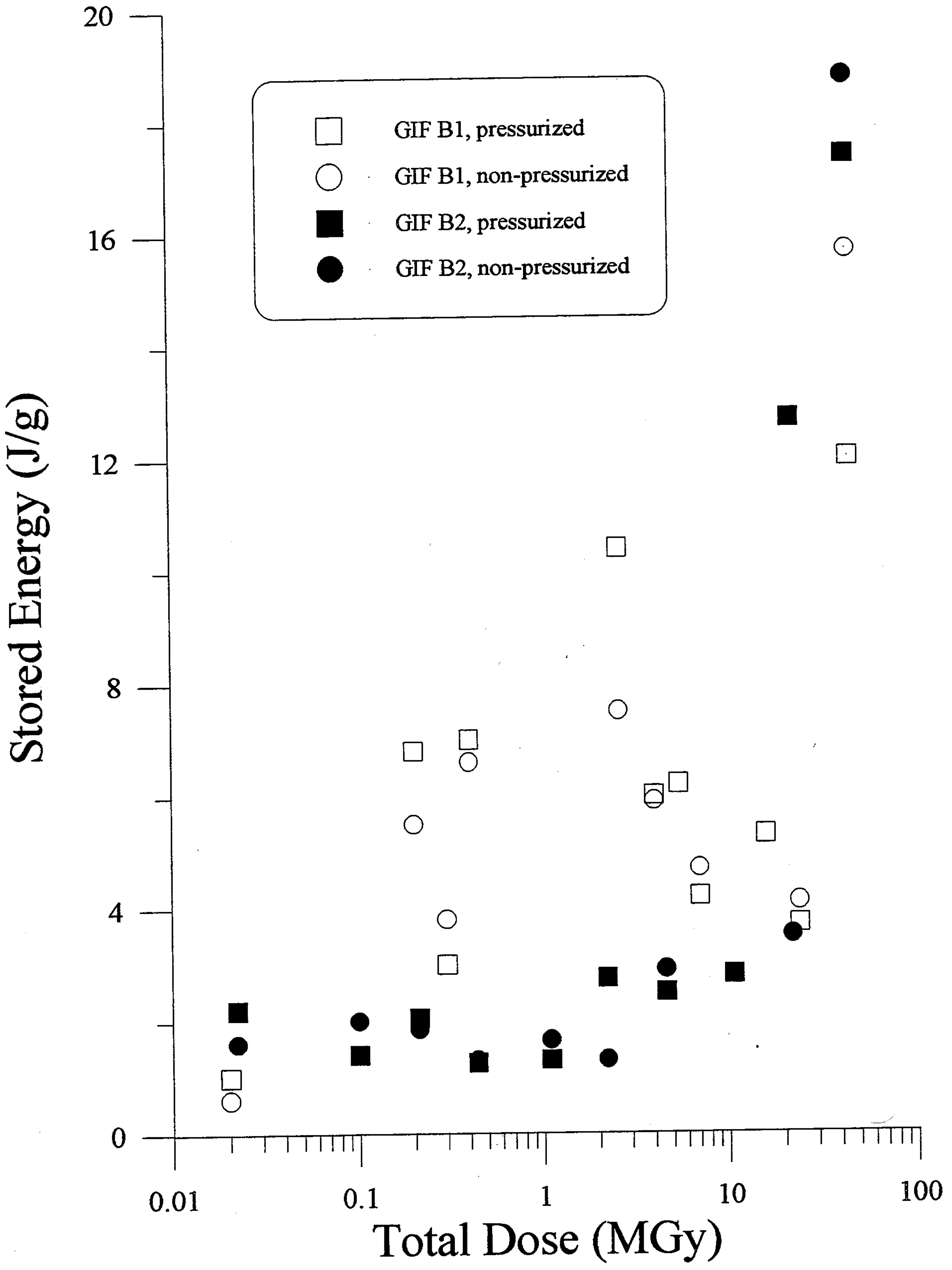

Figure 3: *Measured stored energy as a function of total dose for Harshaw crystals irradiated in the GIF B1 and B2 experiments.*

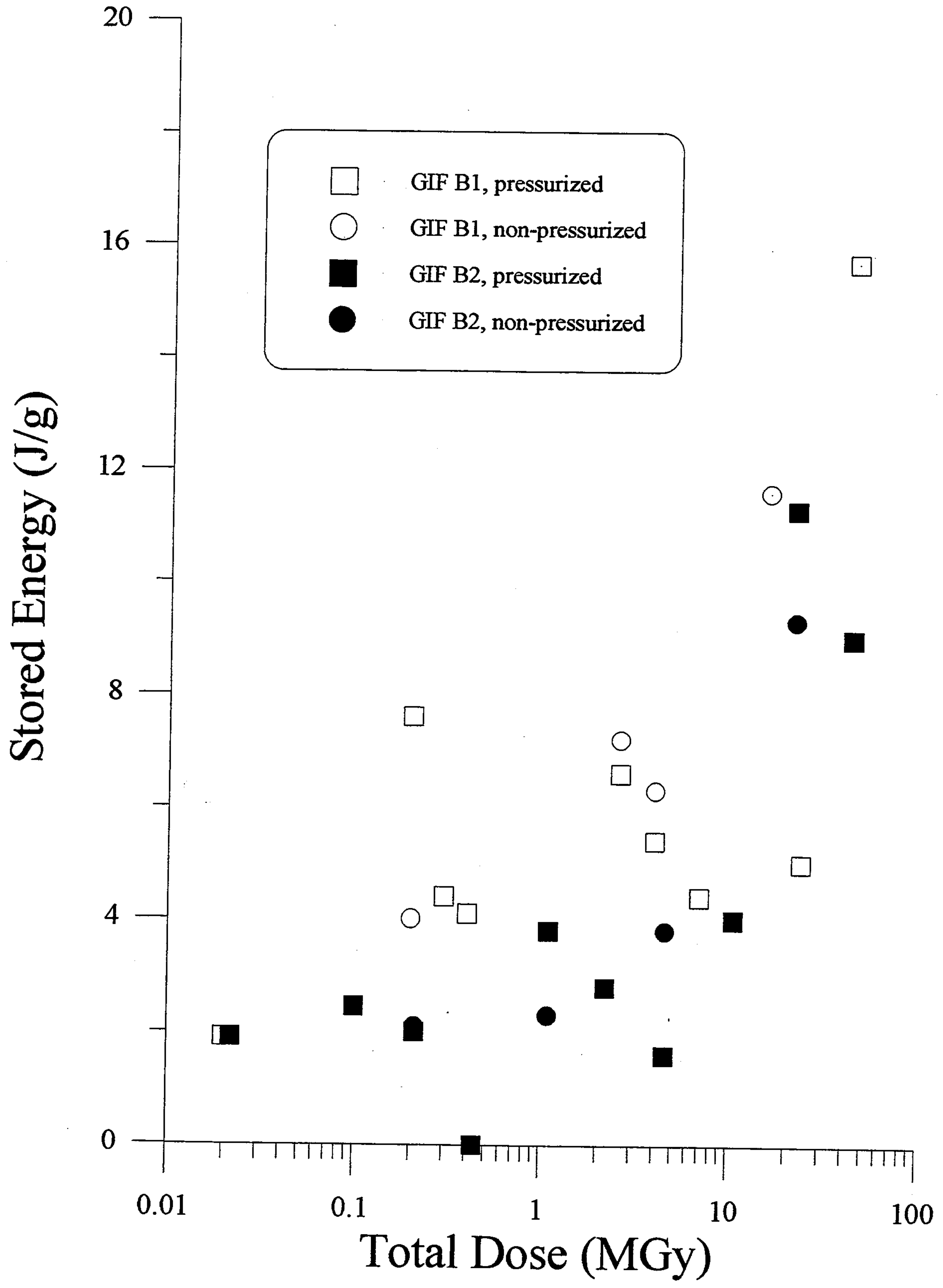

Figure 4: *Measured stored energy as a function of total dose for Sp–800 samples irradiated in the GIF B1 and B2 experiments.*

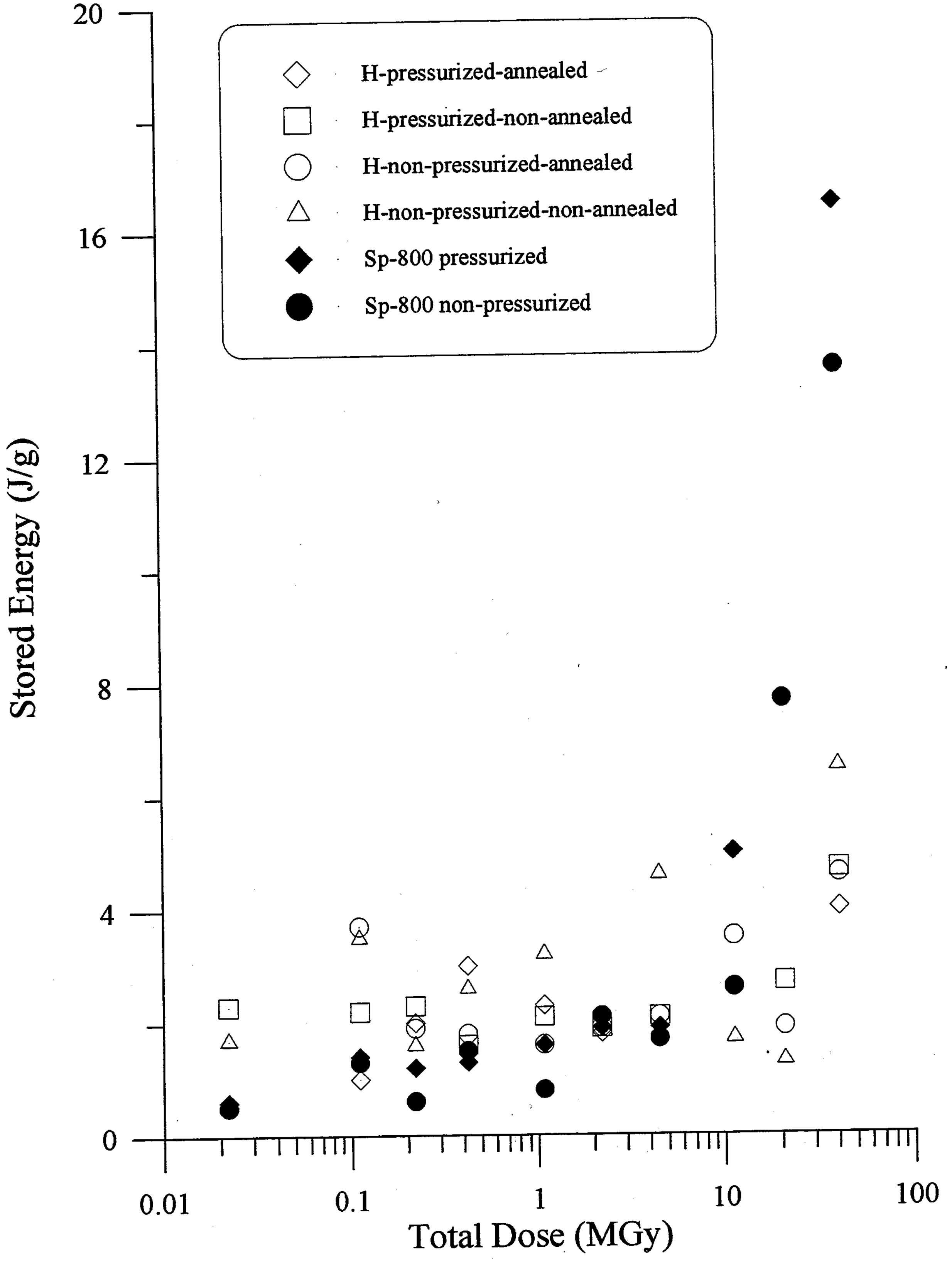

Figure 5: *Measured stored energy as a function of total dose for salt samples irradiated in the GIF B3 experiments.*

could be found as decorated patterns on the surface of the salt samples [Garcia Celma and Donker, 1994a]. Moreover we observed a dark blue rim at the surface of our samples in the thin sections. Although we have some indications of an enhanced damage at the surface of the samples, further research is necessary to prove our suggestions.

For the samples irradiated in the GIF B2 experiments, the results of the stored energy measurements at low total dose are similar to those obtained for the samples irradiated in the GIF B3 experiments. At low total doses there is an approximately constant stored energy at a value of 2 J/g. Above a total dose of 10 MGy this stored energy starts to increase. The maximum stored energy value observed at 44 MGy total dose is approximately 20 J/g.

The GIF B experiments results show that, in general, when natural samples and pure Harshaw monocrystals are subject to the same irradiation experiment, the natural samples contain the same or lower amounts of stored energy than the Harshaw crystals.

Nucleation of colloids is shown to occur at higher total doses in Harshaw monocrystals than in polycrystalline samples. This is probably caused by the different brine content of the samples, their different dislocation density and/or the smaller size of individual crystals in the natural rocks. Notice that colloids and stored energy are two different measures of damage.

In general, the most damaged pieces of NaCl from natural rock salts were found to contain equal or less stored energy than the pure undeformed Harshaw monocrystals irradiated simultaneously. This contradiction with theoretical predictions is, with the help of microstructural criteria, inferred to be due to the fact that the theories do not take dislocation kinetics into account [García Celma et al. 1993b and García Celma and Donker, 1994b].

The bleaching out of colloids in the interior of the crystals was shown to be coupled to the development of intracrystalline recovery microstructures. These creep (recovery) substructures are shown to develop due to irradiation both in samples that were irradiated pressurized and non–pressurized [García Celma and Donker, 1994b].

At minor irradiation doses the presence of brine was found to enhance radiation damage development, but at higher doses brine was found to enhance anneal. This is related to dislocation motion and rearrangement. OH^- ions as lattice impurities are inferred to enhance dislocation motion.

It was already known that Fluid–Assisted Recrystallization (FAR) completely anneals the crystals affected by it [García Celma et al.,1988]. Our experiments show that FAR can take place in irradiated salt without decomposing the brine. They also show that FAR can continuously take place under irradiation, and that it can take place even if the brine is in the form of water vapour. Samples of which the amount of brine measured elsewhere by TG did not surpass the 0.02 weight%, suffered from fluid–assisted recrystallization. The theory of recrystallization has been modified in some points [García Celma et al.,1993a].

Qualitative optical microscope observations on thin sections performed on the irradiated samples sufficed to show that the ratios between recovered/damaged areas and fluid–assisted recrystallized/damaged areas grow with decreasing dose rate.

5. MODEL DEVELOPMENT RESULTS

The first model describing the formation of radiation damage in alkali halides was developed by Jain and Lidiard [Jain and Lidiard,1977]. This model was later modified by Van Opbroek and den Hartog [Van Opbroek and den Hartog, 1985], according to a proposal of Lidiard [Lidiard, 1979]. This modification, the inclusion of a backreaction, was introduced in order to be able to explain the experimental results of Jenks and Bopp [Jenks and Bopp, 1974 and 1977 and Jenks et al., 1975]. A disadvantage of this modified model still is that it does not describe the nucleation stage of the colloids and dislocation loops. Also the effects of impurities, strain and grain boundaries are not taken into account.

The model has therefore been modified to include the processes of nucleation of colloids and dislocation loops [Soppe, 1993]. Two nucleation processes of colloids are taken into account now, homogenous and heterogeneous nucleation. The nucleation of dislocation loops is linked to the existence of impurities. This latest version of the model is capable of reproducing the enhancement of the efficiency of damage formation by impurities. However, just as with the Jain Lidiard model, this model predicts a much higher saturation level for our GIF A experiments than experimentally observed. This is primarily due to the fact that to many parameters in the models are not known with great accuracy. Before quantitative comparisons can be made more accurate information on these parameters has to become available [Donker et al, in prep].

Also calculations have been made including the existence of a dislocation concentration gradient to better simulate the heterogeneities needed for the (enhanced) nucleation of colloids [García Celma and Soppe, in prep].

It is possible to improve the existing models incorporating the newly gained insigths and following a quantitative validation process which could lead to a careful estimation of the unknown and non–experimental parameters in the models, however this would require a lot of additional research work, which unfortunately cannot be performed at this moment. Anyway the models seem qualitatively correct since they can reproduce the trends of all the observed phenomena and, although they can never be absolutely tested, all the calculations performed with them yield an overestimation of the damage obtained in our GIF A experiments (up to high total doses).

6. DISCUSSION AND PARTIAL CONCLUSIONS

6. 1. Dose rate effects (thesis a)

The statement that *low dose rates* are more efficient than high dose rates in producing damage for the same total dose holds for given irradiation condition intervals, sample compositions, and microstructures. However, it does not hold for long times of experimentation in polycrystalline material, and the modified Jain–Lidiard model (1985) exaggerates the efficiency of low dose rates.

Moreover, to reach high doses at low or moderated dose rates, long periods of time are necessary and the subsequent increasing importance of crystal creep processes and its coupled anneal increasingly hinder radiation damage development, at least when the experiments are performed at 100°C.

6. 2. Plastic deformation and impurities effects (thesis b)

Plastic deformation (creep) of crystals takes place due to dislocation development and motion. Natural crystals are always deformed to a certain extent. Dislocations fixate colour centres, thus enhancing nucleation of Na–colloids during irradiation of the crystals that contain them. However, during irradiation (both in pressurized and non–pressurized samples) creep takes place and the motion of dislocations involves motion and anneal of colloids. Since NaCl creeps and adopts another microstructure quickly (it is said to have a fading fabric memory), the starting state of deformation of the samples becomes irrelevant for long experiments. Therefore, the enhancement of colloid nucleation caused by strain in natural NaCl crystals as compared to the pure undeformed single crystals of the theories is only valid for relatively short experimentation times, i.e. either low total dose or very high dose rate experiments. The enhancement of damage by deformation is probably irrelevant for long periods of time and low dose rates.

Nonetheless, the existence, and therefore the development of new grain boundaries affects radiation damage stabilization, at the microscopic level. The grain and subgrain boundaries are the most efficient diffusion paths in the samples, and the differences in concentration of mutually annihilating defects which arise from the different diffusivities towards these big diffusion channels where they all disappear, are at the origin of damage heterogeneity and consequent stabilization.

Regarding chemical crystal defects, such as *lattice impurities*, only the effect of brine as lattice impurity has been evidenced by the experiments. OH^- ions as lattice impurity, ease Na–colloid nucleation, but also seem to enhance anneal at longer periods of time. This is probably due to enhanced dislocation mobility produced by OH^- ions as lattice impurity.

Since huge amounts of time are involved to reach large total doses at low dose rates, the effect of both interstitial brine and dislocations is that they reduce the efficiency of irradiation in damaging the rock salt for increasing times (and total dose) at the same dose rate. This also implies that there is a natural limit to the enhancement of damage caused by low dose rates, since at a given dose rate the time needed to produce damage has to be longer than the time needed for creep in natural situations.

Intracrystalline creep by irradiation has been more intense in the natural rocks than in the pure single crystals of NaCl. This justifies that the most damaged parts of natural crystals do not contain as much stored energy as the pure undeformed single crystals, at least for long experiments. The difference in creep properties has been attributed by us to the different density of preferrent diffusion paths between these two sorts of samples. The diffusion path network is denser in natural rocks which contain many grain and subgrain boundaries.

6. 3. Saturation of damage in natural rock salt (thesis c)

For the Sp–800 samples irradiated in GIF A, *damage saturation* has been reached at 140 J/g, or in other words, in the conditions of our experiments only less than 2 mol% of the natural salt can be decomposed by irradiation. This saturation of damage is also predicted by the last versions of our models for pure undeformed NaCl, however, our experiment shows that saturation also occurs in natural rock salts. Note that 12 mol% decomposition is the lower limit required for spontaneous sudden back reactions taking place.

Radiation damage saturation, as all saturations phenomena, has to be described as due to an enhancement of the back reaction with increasing total product of the forward reaction (damage). It is therefore dependent on the concentration of defect present and on the rate of production of new defects, and thus in our case it ought to be dependent on the dose rate. An important difference between our experiments and other experiments in which much higher damages were reached is that in these last experiments the dose rate was about a factor 20 higher than in our experiment. The processes which could bring about this enhanced back reaction are discussed in 6.4.

It is important to emphasize in our GIF A experiment the saturation of damage cannot be assumed to be due to fluid assisted recrystallization. The duration of the experiment was such that it has to be assumed that at the moment in which saturation of damage was reached the brine present at the start of the experiment had already been consumed. Note that in this experiment the rate of colloid development at the start of the experiment was higher than the rate of fluid assisted recrystallization. Fluid assisted recrystallization consequently took place on crystals which had already developed colloids and brine was decomposed, possibly even completely decomposed after various fluid assisted recrystallization episodes. This is confirmed by the fact that during the DTA measurements of the samples irradiated to high total doses no mass loss is observed, contrary to samples irradiated to low total doses where mass loss due to the evaporation of water has been observed. This would mean that even in absence of H_2O damage saturation is reached.

6. 4. Explosive vs. gradual back reactions (thesis d)

The saturation of radiation damage must be due to an increased rate of anneal mechanisms at high damage levels. Whether this increasing rate of the back reaction is only due to the shorter diffusion distances which have to be surpassed by the F– and H–centres in order to meet an annihilating defect, or is also due to an increasing number of creep mechanisms, is not yet clear.

The behaviour of rock salt creeping as a consequence of irradiation has never been extensively studied. However, the creep behaviour of salt when the damaging agent is stress is well known. Comparing the behaviour of creep with that of radiation–induced creep, which is the same at the microstructural level, it can be doubted whether the reached damage saturation stage will be stable or transitory. Indeed, this analogy suggests that damage (and its stored energy aspect) could increase again after a given total dose has been reached.

In 'normal' creep high strain rates produce brittle failure while low strain rates induce steady state at stress levels which are lower the lower the strain rate. Extrapolation of this analogy, on the basis of the microstructural observations performed [García Celma and Donker, 1994b] would imply that:

a) high dose rates will not produce damage saturation but brittle failure of the samples, perhaps the 'explosive back reactions' observed at the Groningen University [Den Hartog, 1988; Den Hartog et al., 1990; 1992; 1993a, and 1993b] and,

b) low dose rates, as those expected for a repository would induce damage saturation at lower damage levels.

Anyway, even assuming that the observed saturation of damage would be a transitory state, it has to be concluded that this saturation would also have taken place if the experiment had been performed with repository relevant dose rates. This last conclusion is based on the fact that the total dose reached in our GIF A experiment (1223 MGy) is higher than that of a radioactive waste repository (276.6 MGy) [De Haas and Helmholdt, 1989], and therefore the damage can not increase more.

6. 5. Intra and intercrystalline creep mechanisms (thesis e)

Regarding *intercrystalline* processes, *fluid–assisted recrystallization* was already known to eliminate radiation damage. In our work fluid–assisted recrystallization has been proven to be able to take place in irradiated polycrystalline salt without decomposing the brine, as long as Na colloids have not yet extensively developed. It has also been shown that fluid–assisted recrystallization can occur if the brine is present as vapour, and that recrystallization can continuously proceed even on already recrystallized areas. If, before colloids can develop into a crystal, the crystal is cleaned of defects (F– and H–centres) by recrystallization, brine will never be consumed and colloids will never develop.

In short, for lower dose rates, the ratios of anneal by fluid–assisted recrystallization /damage development, as well as the ratios of anneal by Intracrystalline creep/damage development increase. Therefore, in spite of the enhancement of damage for low dose rates it follows that for each low dose rate (e.g. lower than 100 kGy/h) there must be a different and lower saturation level of radiation damage (lower than 2 mol%.). This could be proven by experiments as those performed in GIF A, but with lower dose rates, e.g. dose rates starting at 100 kGy/h.

7. CONCLUDING REMARKS

In the mid eighties some experimental results had been interpreted as meaning that it could not be excluded that very high radiation damage effects (the upperboundary would be of about 20 mol % decomposition of the rock salt) could be built up in a repository. The value of 20 mol% corresponds with the percolation limit for a simple cubic lattice. Later on it was recognized

that, since all the damage is build up in the chlorine sublattice, which is face centered cubic, the percolation limit for this sublattice should be used. This results in a upperboundary of about 12 mol% decomposition of the rock salt.These high percentages of damage can lead to a sudden back–reaction accompanied by a heat pulse and crack development in the rocksalt around the High Level Waste boreholes. As pointed out in the problem definition at the beginning of this article, in order to control the veracity of these proposition we had to rely on mathematical model predictions since the active life of a repository is much larger than that of a research worker. When planning the research we had to choose for the experimental part whether to reproduce the total dose to be expected in a repository by using unrealistic high dose rates, or to reproduce a repository relevant dose rate and never reach the total dose (or almost both , what we in fact did). This experimental work, however requires long lasting experimentation.

Therefore in the framework of the Dutch research program (OPLA) for radioactive waste disposal, a twofold approach was chosen to tackle the problem : one of the research approaches assumed that the upperboundary could be reached and tried to calculate the consequences, the other research approach tried to know whether it is reasonable to admit the possible existence of such an amount of damage as assumed in the upperboundary.

This twofold approach resulted in three research actions which were started simultaneously. The two last research actions, which regarded the second approach, constituted the basis of the research on radiation damage of the HAW–project, while the first approach action was undertaken outside the framework of this project [Prij, 1991]. Anyway, in order to round up the discussions on this problems, Dr. Prij has been as kind as presenting two contributions to this report which clarify the upperboundary approach and the results of it, and constitute article 2 and 23 of this volume.

The three research actions can be resumed as follows:

a) the first action, regarding the upperboundary approach, consisted of assuming that the stored energy in the solid rocksalt steadily accumulates without ever reaching a saturation. Instead of a damage saturation, when a given percolation limit is reached (assumed to lie at 12 mol % decomposition of the salt) all the stored energy could be released in an explosive back reaction. The consequences of explosions in the containment of the waste were then studied for values of suddenly released stored energy which were larger than that corresponding to the 12 mol% of salt decomposition. The result of this analysis was rather reassuring. For the type of repository considered [Prij, 1995a], the effects of such a back reaction were calculated to be rather limited [Prij ,1991; Prij,1995b]. Notice however, that in order to have the explosion, radiation damage has to be at least as high as 12 mol% decomposition of the salt.

b) the second action, consisted in elucidating whether radiation damage in natural rocksalt reaches a saturation or goes on accumulating until a percolation limit is reached, as assumed in action *a* . As described in this work (see as well Figure 2) radiation damage saturates in our experiments at less than 2 mol % of salt decomposition, and this is evidently not enough for spontaneous back reactions taking place.

c) the third action consisted of long lasting experiments of irradiation under conditions as near as possible to those expected in a repository (although up to lower integrated doses, of course) and comparison of the results with these given by mathematical models. After implementing the nucleation of colloids, the effect of impurities, and the effect of dislocation densities and evolution in different versions of the models, the models are now able to reproduce even effects which were considered odd, e.g. the impurities effect and the blue–white grain boundary phenomenon. Anyway, although the models do qualitatively reproduce all the observed phenomena, all the model calculations which have been performed in the framework of this project yield an overestimation of the damage (see as well Fig. 2). This could be due to the fact that the models do not yet include the effect of creep processes which anneal the stored energy and which have been shown to be active during and due to irradiation in natural rock salt.

It could be argued that low dose rates would nonetheless enhance damage development and thus produce a higher saturation level than that obtained by us. However, notice that the dose rate for which our GIF A experiment was performed produce saturation and that the most important difference in the irradiation conditions between our experiments and previously reported experiments with "explosions" [Groote and Weerkamp 1990] is that our mean dose rate was lower than that used in the experiments where brittle failure of the samples (explosions) occurred, and that our samples are natural polycrystalline rock salt while the "exploded" samples were synthetic monocrystals containing unnaturally high amounts of potasium.

The obtained saturation of damage can only be due to a balance between the rate of anneal and the rate of damage development. We assume that in the work at high dose rate where the explosions have been reported, the rate of anneal is slow enough to go unnoticed since there are very high damage production rates. But we have shown in our work that damage production rates lower than those used in the experiments of the explosive back reactions already have to compete with anneal and reach a saturation compromise (GIF A). We can therefore conclude that lower damage production rates in natural rocks will probably encounter a harder competition from the anneal processes and result in lower levels of stored energy at saturation. Whether this increased importance of anneal at low dose rates in natural rocks does always have a dislocation dynamics support (is due to creep, as observed in GIF B) or is mainly due to diffusion gradient development depending on the natural rock salt heterogeneity (as also observed in GIF B and simulated with the models) or, most probably both, we are not able to say.

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Turkin, Vainshtein
& den Hartog
Nucl. Instr &
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vol 153, pages
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A drawback of our experiments has been that they have only been performed at one irradiation temperature (i.e. 100 °C). This limitation stems from the fact that the laboratory experiments were originally only planned to monitor the HAW field experiment. In the HAW experiment the effect of temperature was planned to be studied. Unfortunately this experiment has been cancelled. The temperature of 100 °C was chosen because this temperature had been reported by other researchers as the temperature where the efficiency of damage formation was maximal. The Jain–Lidiard model, however, predicts that this temperature of maximal efficiency for damage formation is highly dose rate dependent and that it decreases with decreasing dose rate. We, therefore, have to consider the effects of dose rates and /or temperatures which are lower than in our experiment (it may be obvious that higher dose rates and/or temperatures will only result in less damage).

If we had performed our GIF A experiment at a lower temperature the efficiency of damage formation at low total doses, as shown in other experiments [Levy et al., 1981; Den Hartog, 1988], would have been lower, since the saturation level of the damage is, according to theoretical models [Van Opbroek and den Hartog, 1985; Soppe, 1993], expected to be higher than that in our experiment, it follows that this saturation level can only be reached at a higher total dose than that for which it was reached in our experiment. Since in our experiment the saturation level is reached at a total dose which is higher than the maximal total dose expected for a repository (800 (see figure 2) vs. 277 MGy [Prij, 1995a] respectively) a lower temperature alone will not lead to damage levels that are higher than the saturation level of 140 J/g found in our experiments.

In the previous sections we have discussed that the effect of dose rates lower than that used in our experiment will be an enhancement of the damage formed at low total doses, however the saturation level will not only be lower but also be reached at a lower total dose. The effect of a lower dose rate alone cannot either be the obtainment of damage levels that are higher than the sturation level of 140 J/g found in our experiments.

However, in a repository, there will always be a trajectory in which both the dose rate and the temperature are lower than in our experiments (see [Prij, 1995a]). The effect that these conditions will have on the efficiency of damage formation and thus on the amount of damage formed is difficult to predict without the use of a theoretical model. The analyses performed with the extended Jain–Lidiard model [Soppe et al., 1994; Prij, 1995b] show that the time period in which there is a considerable efficiency for damage formation is restricted to the first 100 years of disposal for all the conceptual repository designs considered. After 100 years of disposal a stationary damage level that is not higher than 300 J/g is reached. At this moment there is no scientific basis to assume that the values predicted by this model will be an underestimation. In fact we have many arguments that the opposite, an overestimation, is very propable. Since, although for some low dose experiments the model predicted lower damage levels than experimentally observed, in all comparisons between this model and experimental results obtained for high total doses (> 22 Mgy), the model predicted higher stored energy values than experimentally observed. Also we have observed anneal mechanisms in our experiments which are not yet included in the model (see paragraph 6.5.). We have already discussed that these anneal mechanisms will grow in importance with decreasing dose rate. Therefore, for a repository lower damage levels than predicted until now may be expected.

And now, a last remark regarding repository conditions. In Fig. 2 the possible highest total dose of a repository as regarded at the start of this project has been plotted. This maximum total dose is obtained if the waste is reprocessed 3 years after its retrieval as spent fuel from a reactor, and is consequently placed in an interim storage during 10 years. Please notice that the interim storage duration which is considered nowadays is larger (about 50 years) and will consequently lead to repository total doses a factor two lower than that considered in Fig. 2. It will also lead to lower maximal damage levels than the 300 J/g considered in the previous paragraph. This means that, even in the absence of fluid assisted recrystallization the damage will be very low. Now, if we moreover consider, as we have also shown in this work [García Celma et al., 1993a], that the combination of fluid assisted recrystallization and low levels of developed damage can lead to continuously renewed complete anneal of radiation damage we have to conclude that it could very well be that the damage development in a repository will never surpass the colloid nucleation stadium.

8. ACKNOWLEDGEMENTS

The collaboration between the Dutch (OPLA) and Spanish (ENRESA) state organizations which made this work possible takes place in the framework of the European Communities R&D Programme on Management and Storage of Radioactive Waste. This work has been performed under contract Nº CEC F1-1W-0235-E(TT).

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